Nature of crossover from classical to Ising-like critical behavior

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We present an accurate numerical determination of the crossover from classical to Ising-like critical behavior upon approach of the critical point in three-dimensional systems. The possibility to vary the Ginzburg number in our simulations allows us to cover the entire crossover region. We employ these results to scrutinize several semi-phenomenological crossover scaling functions that are widely used for the analysis of experimental results. In addition we present strong evidence that the exponent relations do not hold between effective exponents.

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It is now well established that the critical behavior of large classes of systems—including uniaxial ferromagnets, binary alloys, simple fluids, fluid mixtures, and polymer blends—belongs to the three-dimensional (3D) Ising universality class. However, this behavior is only observed asymptotically close to the critical point, whereas *classical* critical behavior is (sometimes) observed at temperatures farther from the critical temperature T_c, before one enters the noncritical background. At intermediate temperatures, a continuous crossover occurs from one universality class to the other. Due to the limited extent of the asymptotic (Ising) regime, the thermodynamic behavior in this crossover region is extremely relevant from an experimental point of view and hence has attracted long-standing attention. Despite this attention, several fundamental questions concerning the crossover are still open to debate.

First, we note that the vast majority of all relevant studies is limited to the one-phase region only. The behavior of the susceptibility (compressibility) in this region is described by means of several approaches, which all consist of more or less phenomenological extensions of the renormalization-group (RG) description of the critical behavior. In Ref. [1] three different descriptions have been compared. From a nonlinear treatment of ϕ^4 theory at fixed dimensionality $d = 3$, Bagnuls and Bervillier [2,3] obtained functions describing the full crossover from asymptotic critical behavior to classical critical behavior. Belyakov and Kiselev [4] carried out a first-order ε-expansion, which was then phenomenologically extended to yield the correct asymptotic behavior in the Ising regime. Finally, Ref. [1] discusses an extension by Chen *et al.* of the work of Nicoll and Bhattacharjee [5], based on an RG matching technique. All three approaches yield rather similar results and suggest (for simple fluids) a smooth monotonic crossover.

Yet, from the experimental side it has not been so easy to confirm this picture or even to make judgments concerning the quality of the various predictions. Widespread attention attracted the findings [6] on micellar solutions, in which exponents were observed that did not fit into any of the known universality classes. This was analyzed by Fisher [7] in terms of a crossover effect, suggesting that a nonmonotonic variation of the susceptibility exponent in the one-phase region is an intrinsic property of the universal crossover scaling function. This possibility of nonmonotonicity was then essentially confirmed in Ref. [8], although again an empirical extension of RG theory had to be invoked as well as extremely strong higher-order corrections. All authors stressed the need for much more accurate experimental results and explicit calculations of the crossover behavior. Indeed, several experiments have been carried out on polymer blends [9,10], which were analyzed in terms of the crossover solution by Belyakov and Kiselev. Polymer systems offer the advantage that the Ginzburg number ruling the crossover can be varied by varying the length of the polymer chains. Nevertheless, it proved difficult to cover the full crossover region and results for different polymer blends with widely varying chemical properties had to be combined. This in turn led to conjectures concerning an unexpected pressure dependence of the Ginzburg number G, which needed to be fitted to each system separately [10]. A determination of effective exponents has not been attempted in these studies. More recently, Anisimov and coworkers [11] focused on the possibility of nonmonotonic variations of effective exponents in complex fluids, whereas also for polymer solutions a sharp, nonmonotonic crossover of the susceptibility exponent has been reported [12]. However, it is unclear how the results on micellar solutions fit into this picture and also the applicability of the crossover solution by Chen *et al.* to systems which exhibit a nonmonotonic crossover is subject to some debate. Nonuniversal crossover (depending on parameters in addition to the Ginzburg number) is suggested [11], but this behavior may be particular to complex fluids due to the occurrence of mesoscopic lengths in addition to the correlation length.

In Ref. [13], we presented a numerical study of the crossover to classical critical behavior on either side of the critical temperature in *two-dimensional* systems. This revealed a strictly monotonic crossover of the susceptibility exponent in the disordered phase (one-phase region) and a nonmonotonic variation in the ordered phase. Although this was an interesting finding in itself, it also stressed the importance of a study of 3D systems, since

only these systems can be compared with the various theoretical crossover descriptions and also an even *qualitative* difference with the two-dimensional case could not be excluded. It is the objective of this paper to present the results of a major numerical effort to determine the crossover in 3D spin systems. These results allow, for the first time, a detailed and rigorous test of the above-mentioned crossover functions.

The crossover is ruled by the parameter t/G, where $t \equiv (T - T_c)/T_c$ is the reduced temperature and G the so-called Ginzburg number. Asymptotic critical behavior occurs for $t/G \ll 1$ and classical critical behavior is expected for $t/G \gg 1$. The additional requirement that t must be small implies that only systems with a very small G (large interaction range) allow an observation of the full crossover. In simple fluids, e.g., the crossover is never completed before leaving the critical region. Numerical calculations offer the advantage that G is known precisely and can be made arbitrarily small by increasing the range of the interactions. Thus, we could vary t/G over more than eight orders of magnitude, compared to four orders of magnitude in experiments on polymer blends [9]. The large variation of t/G requires the simulation of systems with very large coordination numbers. Until now, this constituted the main bottleneck for explicit calculations. However, the advent of a new Monte Carlo algorithm [14] for long-range interactions has now allowed us to cover the full crossover region.

We have simulated classical spin systems, consisting of 3D simple cubic lattices with periodic boundary conditions. The systems are described by the Hamiltonian $\mathcal{H}/k_BT = -\sum_{ij} K_d(\mathbf{r}_i - \mathbf{r}_j)s_is_j$, where $s = \pm 1$, the sum runs over all spin pairs, and the coupling depends on the distance $|\mathbf{r}|$ between the spins as $K_d(\mathbf{r}) = cR_m^{-d}$ for $|\mathbf{r}| \leq R_m$ and $K_d(\mathbf{r}) = 0$ for $|\mathbf{r}| > R_m$. For any finite R_m, the critical behavior of this model belongs to the Ising universality class, but for $R_m \to \infty$ it will be classical. The consequent singular dependence on R_m of all critical amplitudes has been derived in Refs. [15,16]. In order to avoid lattice effects we formulate our analysis in terms of an *effective* interaction range R [15]. The R dependence of the Ginzburg number is given by $G = G_0R^{-2d/(4-d)}$, such that the crossover occurs as a function of tR^6/G_0. By increasing R we can reach the classical regime while still keeping $t \ll 1$ in order to stay within the critical region. On the other hand, very small values of t imply a strongly diverging correlation length and extremely large system sizes are required to avoid finite-size effects. Thus, we have constructed all crossover functions by combining the results for systems with different Ginzburg numbers, such that t had to be varied only within a limited range. Systems with linear size up to $L = 160$ (ca. 4 million spins) have been simulated, in which each spin interacts with up to 8408 neighbors, corresponding to an effective interaction range of 9.8 intermolecular distances.

We first consider the experimentally most widely studied case, *viz.* $T > T_c$. We have calculated the susceptibility from the magnetization density m, $\chi' = L^d\langle m^2\rangle/k_BT$, and the scaled susceptibility $\tilde{\chi} = k_BT_c(R)\chi'$. In the Ising limit, the latter quantity diverges as $C_I^+ t^{-\gamma}$, with $\gamma = 1.237$ [17] and $C_I^+ = 1.1025$ [18], whereas mean-field theory predicts $\tilde{\chi} = 1/t$, i.e., $\gamma_{\rm MF} = 1$. The leading range dependence of the critical susceptibility amplitude is given by $R^{2d(1-\gamma)/(4-d)}$ [15,16]. Thus, if one plots the data as a function of the reduced variable t/G, a data collapse should be obtained for $\tilde{\chi}G_0/R^6$. We will now test whether this quantity reproduces the predicted crossover behavior and how well it is described by various theoretical expressions. First, we consider the phenomenological generalization of first-order ε-expansion results obtained in [4]. The main significance of this function lies in its rather widespread use for the analysis of crossover effects in polymer systems. It is given in the following form,

$$t/G = [1 + \kappa(\tilde{\chi}G)^{\theta/\gamma}]^{(\gamma-1)/\theta} \times \left\{ (\tilde{\chi}G)^{-1} + [1 + \kappa(\tilde{\chi}G)^{\theta/\gamma}]^{-\gamma/\theta} \right\} , \qquad (1)$$

where $\theta = 0.508$ (25) [19] is Wegner's correction-to-scaling exponent and $\kappa \approx 2.333$ a universal constant. Asymptotically close to the critical point, $\tilde{\chi} = G^{\gamma-1}(1 + \kappa^{\gamma/\theta})^{\gamma}\kappa^{-\gamma/\theta}t^{-\gamma}$ and for $t/G \gg 1$ $\tilde{\chi}$ exhibits the limiting behavior $1/t$. As the curve reproduces the amplitude of the mean-field asymptote, the only remaining adjustable parameter is a multiplicative constant in G. This is precisely the way G is determined in experimental analyses. Considering $\tilde{\chi}$ given by (1) as the master curve, we set $G_0 = [C_I^+\kappa^{\gamma/\theta}(1 + \kappa^{\gamma/\theta})^{-\gamma}]^{1/(\gamma-1)} \approx 0.1027$ such that the 3D Ising asymptote coincides with the curve in the limit $t/G \ll 1$. On the other hand, we may also calculate the *exact* Ginzburg number from the expression given in Ref. [4], which for our model reduces to $G = 27/(\pi^4 R^6) \approx 0.27718/R^6$. The former value $G_0 = 0.1027$ leads to a precise reproduction of the Ising asymptote and hence has been used in the graph of the crossover curve, but the exact value of G may yield a better description of the overall crossover behavior. Secondly, we consider the crossover function resulting from the nonlinear RG treatment of Ref. [2]. It is presented in terms of a phenomenological function $\chi^*(t/g_0^2)/g_0^2$, which represents the RG calculations with a relative error of less than 10^{-4} [2]. The merit of this field-theoretic treatment is that it has a more solid foundation than Eq. (1). Also χ^* reproduces the mean-field asymptote and thus we adjust the parameter g_0 such that $\chi^*(t/g_0^2)/g_0^2$ coincides with $\tilde{\chi}$ in the Ising regime. As mentioned before, a third crossover function for the susceptibility exists, based on an RG matching technique. However, we have not included this function in the present discussion, because of its very close resemblance to the solution of Ref. [4].

Figure 1 shows our results for the susceptibility in the symmetric phase ($T > T_c$) along with the two theoretical curves. One observes that the simulations accurately reproduce both the Ising and the mean-field asymptote (dotted lines). In between, the data exhibit a gradual crossover, just as predicted by both theoretical

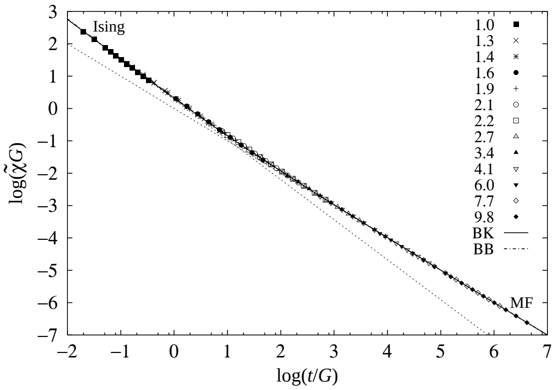

FIG. 1. Crossover curve for the magnetic susceptibility above T_c. The numbers in the key refer to the effective interaction range R. "BK" and "BB" indicate the crossover functions of Refs. [4] and [2], respectively.

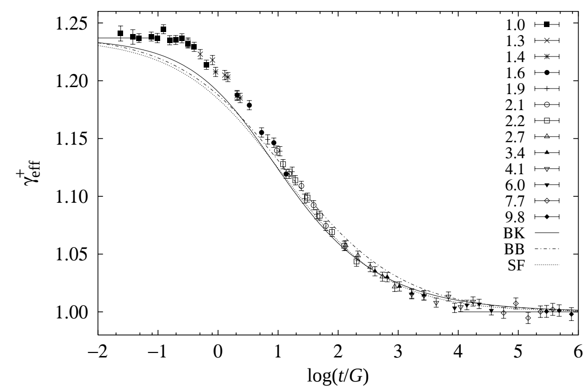

FIG. 2. The effective susceptibility exponent $\gamma_{\rm eff}^{+}$ above T_c along with three theoretical calculations for this quantity. "SF" refers to the first-order ε-expansion of Refs. [20,7].

curves. However, the vertical scale of the graph covers ten decades, which makes it difficult to observe subtle deviations. The logarithmic derivative of the susceptibility, $\gamma_{\rm eff}^{+} \equiv -d\ln\tilde{\chi}/d\ln|t|$, constitutes a much more stringent test, see Fig. 2. The excellent collapse of the data for all our models with widely differing interaction ranges on a master curve suggests that an interpretation of the crossover in terms of a universal crossover function (i.e., described by a single crossover argument t/G) is appropriate here. In this graph, we have also included a third calculation by Seglar and Fisher [20,7], which was shifted along the horizontal axis such as to reproduce the initial deviations of $\gamma_{\rm eff}^{+}$ from the mean-field value. Apart from this somewhat arbitrary shift, the curve resembles the other predictions and is given by

$$\gamma_{\rm eff}^{+} = 1 + (\gamma - \gamma_{\rm MF})E[\ln(t/G)] \;, \tag{2}$$

with $E(\ln y) = 1/(1 + y^{\varepsilon/2})$, where $\varepsilon = 4 - d$. Our results for $\gamma_{\rm eff}^{+}$ display a smooth, monotonic crossover from the Ising value 1.237 to the classical value 1. While this agrees with the various theoretical functions shown, it clearly contradicts the conjecture of Ref. [7], according to which a *nonmonotonic* variation of $\gamma_{\rm eff}^{+}$ in the symmetric phase might be a property of the universal crossover scaling function. The good description of the initial increase of $\gamma_{\rm eff}^{+}$ upon approach of the critical point is an encouraging result, since G has been set to its exact value (see above) in Fig. 2 and hence there is *no* adjustable parameter in the solution (1). However, we note that the figure also reveals a remarkable discrepancy between the theoretical calculations and our results. Namely, after the initial deviation from $\gamma_{\rm MF}$, the actual curve proceeds with a considerably steeper increase than predicted by any of these functions. Following Fisher [7], we define the gradient $\Gamma \equiv -\partial\gamma_{\rm eff}^{+}/\partial\log t$. Indeed, Γ reaches values as high as 0.84 per decade, in contrast to the theoretical functions for which the maximum of Γ lies in the range 0.64–0.74. In practice, this implies that the full crossover is completed *in one to two decades less* than predicted. A final interesting aspect of Fig. 2 is the fact that even for $R = 1$ no significant overshoot of the effective exponent above the Ising value can be observed. This is remarkable, as it is expected [21] that the 3D nearest-neighbor Ising model exhibits a *negative* leading Wegner correction. As the form of the curve makes a further increase of $\gamma_{\rm eff}^{+}$ for smaller values of t rather unlikely, we conclude that the actual effect must be very small.

Now we proceed to the temperature region below T_c. We have calculated the susceptibility from the fluctuation relation $\chi = L^d(\langle m^2\rangle - \langle|m|\rangle^2)/k_BT$. We find that the the simulation data faithfully reproduce both the Ising and the mean-field asymptote and that the crossover curve never deviates far from these asymptotes. The overall graph is very similar to that for $T > T_c$. To our knowledge it constitutes one of the first determinations of the full crossover function in the ordered phase. Indeed, experimentally it is very difficult to measure the coexistence curve and hence the crossover function. Also from the theoretical side very few results exist. Bagnuls and coworkers [22] have carried out a calculation using massive field theory. Just as for $T > T_c$ (see above) they present their results in the form of an approximative continuous function. However, this function is only valid for relatively small values of t/G and does not cover the entire crossover region. Indeed, for t/G large it approaches an asymptote with slope 0.97 instead of the classical value 1. Thus, it is not possible to accurately describe our results with this function. The exponent $\gamma_{\rm eff}^{-}$ following from our data is shown in Fig. 3 and displays several noteworthy features. First, also in the ordered phase no nonmonotonicity can be observed within the statistical accuracy, in contrast with the two-dimensional case. Secondly, the increase of the effective exponent upon approach of the critical point is even faster than for $T > T_c$, with Γ as high as 0.11. While $\gamma_{\rm eff}^{+}$ gradually starts to deviate from $\gamma_{\rm MF}$ when t is decreased, the increase of $\gamma_{\rm eff}^{-}$ is rather abrupt. We view this effect as the precursor of the "underswing" observed for $d = 2$ [13]. In the absence of further calculations of $\gamma_{\rm eff}^{-}$, we have attempted to de-

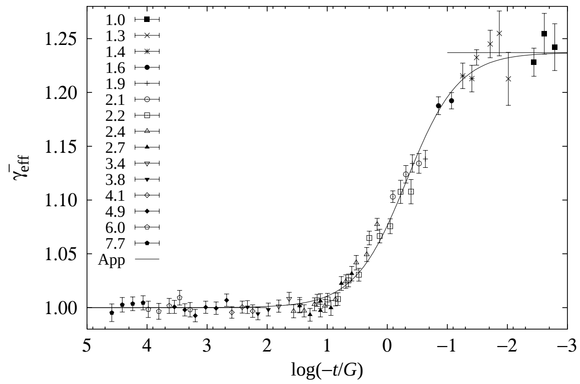

FIG. 3. The effective susceptibility exponent γ_{eff}^{-} below T_c. "App" denotes the approximation discussed in the text.

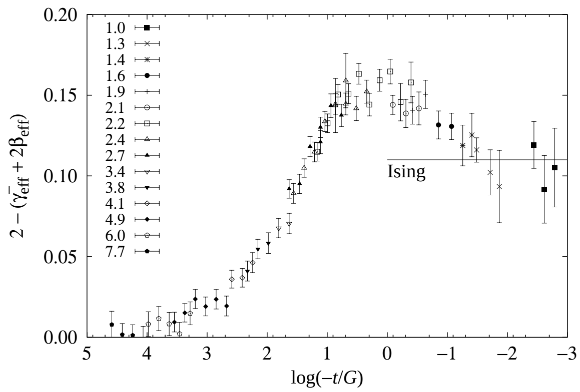

FIG. 4. The quantity $2-(\gamma_{\text{eff}}^{-}+2\beta_{\text{eff}})$ as a function of t/G.

scribe the data by a phenomenological generalization of the exponent crossover function (2). We found that the expression $E(\ln y)=1/(1+y)$ captures the actual behavior reasonably well (see Fig. 3), although the parameter y in the denominator might carry an even larger exponent.

Finally, we have also considered the crossover behavior of the order parameter $\langle|m|\rangle$ for $T<T_c$. The corresponding exponent β_{eff} turns out to vary monotonically from its Ising value 0.3267 to the classical value 0.5. In Fig. 4 we display an interesting consequence of this behavior. Namely, we have plotted the quantity $2-(\gamma_{\text{eff}}^{-}+2\beta_{\text{eff}})$, which should be equal to the effective specific-heat exponent α_{eff}^{-} *if the standard scaling relations hold between effective exponents*. However, this quantity varies between the classical and the Ising value in a strongly nonmonotonic way, which is very unlikely in view of the smooth behavior of γ_{eff}^{-} and β_{eff}. Thus, we consider this as strong evidence for the violation of scaling relations between effective critical exponents. Interestingly, such a breakdown has been inferred from ε-expansions over 18 years ago [23], but could experimentally not be confirmed. For the first time it has now been explicitly demonstrated.

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